



Short communication

Photo-excitation promoted quality factor of carbon nanotubes



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H I G H L I G H T S

- Carbon nanotubes exhibit RCL characteristics in an ac field.
- Quality factor of carbon nanotubes-made devices is measured to be as high as 10^4 .
- Quality factor of carbon nanotubes can be further promoted by photo-excitation.

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Carbon nanotubes exhibit characteristics of resistor–capacitor–inductor in alternating current field and quality factor is estimated to be as high as 1×10^4 . This value can be optically promoted and improvement is verified as a result of photocurrent creation in tubes. Experimental data is supported by *ab-initio* calculations.

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1. Introduction

The quality factor (Q) is a dimensionless parameter often used to characterize damping magnitude of an oscillating system and normally lies on 10^2 – 10^3 for alternating current (ac) driven electronics [1]. Impedance (Z) however emerges as current (I) and voltage (V) oscillate and is expressed as

$$Z = R - j/\omega C + j\omega L \quad (1)$$

where R is resistance, j is imaginary number, ω is angular frequency, C is capacitance and L is inductance; the 2nd and 3rd terms correspond to capacitive (X_C) and inductive reactance (X_L). For resistive circuits, sine wave amplitudes of I and V are always in-phase and imaginary part of Z is zero (i.e. $-j/\omega C = 0$, $j\omega L = 0$) [1–4]. Phase difference occurs as device contains reactive components (i.e. capacitor & inductor) and reactance begins to contribute to Z (i.e. $-j/\omega C + j\omega L \neq 0$). At resonant frequency (ω_0), X_C and X_L cancel each other (i.e. $-j/\omega C + j\omega L = 0$) and energy storage at

capacitor and inductor reaches a maximum. In this case, input power is multiplied by Q expressed as

$$Q = \frac{L\omega_0}{R} = \frac{1}{CR\omega_0} = \frac{1}{R} \sqrt{\frac{L}{C}} \quad (2)$$

Carbon nanotubes (CNTs) are one dimension conductor made of rounded graphene sheets and do not suffer from the peierls distortion at low temperature [5]. Electrical measurements reveal that CNTs also behave as reactive elements in ac field (Fig. 1) and damping originates from curvature induced dipole relaxation and chiral current [6,7]. At low $\omega < 1000$ Hz, Z is mainly contributed by R and both X_C and X_L approximate zero. Spins become polarized as ω increases and X_C begins to contribute to Z . Calculation reveals $X_C = 40$ – 50Ω and X_L due to circulating current lies on 0.1 – 1Ω [8–16]. Studies however lack explanation on energy loss mechanism and Q remains unexplored. In this work, devices that contain a low content of CNTs are made and Q is measured at dark (Q_{off}) and bright conditions (Q_{on}). We find $Q_{\text{on}}/Q_{\text{off}} > 1$ and both Q_{off} and Q_{on} reach a level comparable with that on oxide-based resonators. Optical enhancement has been identified as a result of photocurrent (I_{photo}) creation in nanotubes and is supported by $I_{\text{photo}} \propto Q_{\text{on}}/Q_{\text{off}}$. Photo-carriers however tend to couple with leads; thus create interfacial

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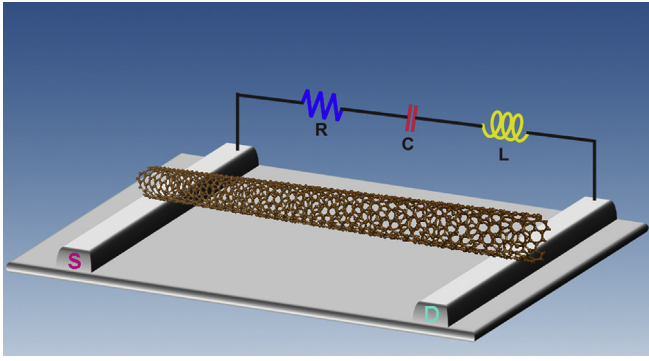


Fig. 1. Scheme of a CNT created RCL circuit.

reactance. Study reveals that I_{photo} also dissipates at tube boundaries and loss mechanism involves Schottky potential (E_{SCH}) induced hopping at metallic (M)/semiconducting (S) tube junctions.

2. Experimental

Arc-discharge produced single-walled CNTs (SWCNTs, 50 mg) are dispersed in $\text{H}_2\text{O}_2/\text{H}_2\text{O}$ solution (1:1 weight %, 100 ml) and oxidation is carried out at 50 °C for 2 h. Oxidized CNTs are deposited onto SiO_2 substrate supported Al electrodes and devices are then placed in a vacuum chamber (10^{-5} torr) equipped with electrical fit-through and laser pointer (632 nm wavelength, spot size 0.5 ~ 1 mm, 5 mW, Horiba-HR800). Compared with single tube, CNT networks produce a large I_{photo} and therefore promote accuracy of electrical measurements [5,8]. The R and I_{photo} are probed with a network analyser (Zentech-3302, ac mode, $\omega = 0.1 \text{ KHz} - 1 \text{ MHz}$, background noise = $\pm 0.5 \text{ nA}$) and measurements are carried out at

various temperatures (T) and bias voltages (V_{bias}). The Q is determined according criteria given by Eq. (2); (i) the R minimum and I maximum (I_{max}) emerge at ω_0 ; (ii) $I \geq 0.707 I_{\text{max}}$ takes place at pass-band width of frequency ($\Delta\omega$) [Fig. 2(a)] [1]; (iii) R is inversely proportional to Q (i.e. $R \propto Q^{-1}$). We first scan devices from 0.1 KHz to 1 MHz to monitor I variation and $\Delta\omega$ is determined by $I \geq 0.707 I_{\text{max}}$. Once $\Delta\omega$ is located the input I is switched from coarse (mA– μA) to fine (μA –nA) modulation to spot I_{max} and ω_0 [Fig. 2(a)]. It is worth mentioning that both SWCNTs and Al-electrodes have a similar work function (4.25–4.28 eV) and do not form an E_{SCH} capable of producing I_{photo} at interfaces [12].

3. Results and discussion

Fig. 2(b) shows $R-\omega$ profiles obtained from three different samples and ω_0 (i.e. R minimum) appears at $170 \pm 20 \text{ KHz}$ for sample-1, $168 \pm 17 \text{ KHz}$ for sample-2 and $166 \pm 18 \text{ KHz}$ for sample-3; the ω_0 variation from sample to sample being the fact that tubes are arranged in various configurations between leads [1,2]. Q however does not change with tube arrangements and is determined only by Z [17]. Fig. 3(a) shows $Q-\omega$ profiles of sample-1 at $T = 300 \text{ K}$ and, the Q_{off} and Q_{on} are calculated according to R profiles at light-off (R_{off}) and -on (R_{on}) [e.g. Fig. 2(b)]. It is clearly that Q_{on} exceeds Q_{off} and $Q_{\text{on}}/Q_{\text{off}}$ approximates 2.1: both Q_{on} and Q_{off} are comparable with that on oxide-based resonators ($\sim 10^4$) [2,3]. The $Q_{\text{on}} > Q_{\text{off}}$, according to Eq. (2), also means an enhanced energy storage at capacitor and inductor (i.e. increased C and L), accounting for ω_0 shift from 181.4 KHz to 176.8 KHz [$\omega_0 = 1/(LC)^{1/2}$, Fig. 3(a)]. Apparently, enhanced C and L originate from I_{photo} creation in CNTs and photo-excitation is further supported by $V_{\text{bias}} \propto I_{\text{photo}}$ [Fig. 3(b)] [18]. Fig. 4(a) and (b) plots C and L at light-off and -on and I_{photo} induced increase is estimated to be 3% and 1.5% for sample-1 (top), consistent with Fig. 3(a). Experiments carried out on sample

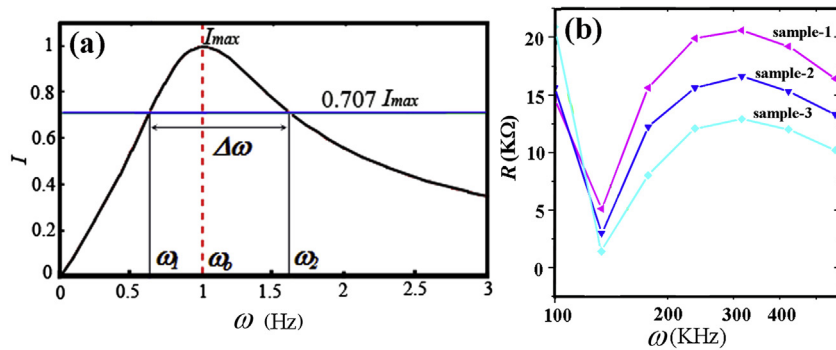


Fig. 2. (a) The relationship of I_{max} with $\Delta\omega$ and ω , (b) and $R-\omega$ plots.

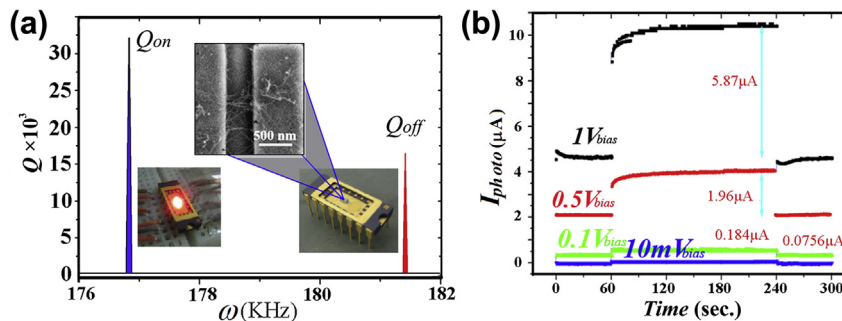


Fig. 3. (a) The $Q-\omega$ profile obtained from sample-1. Insets: SEM image of networked SWCNTs between leads (top), and device at illumination-off (right) and -on states (left). (b) The time evolved I_{photo} profiles of sample-1 at various V_{bias} .

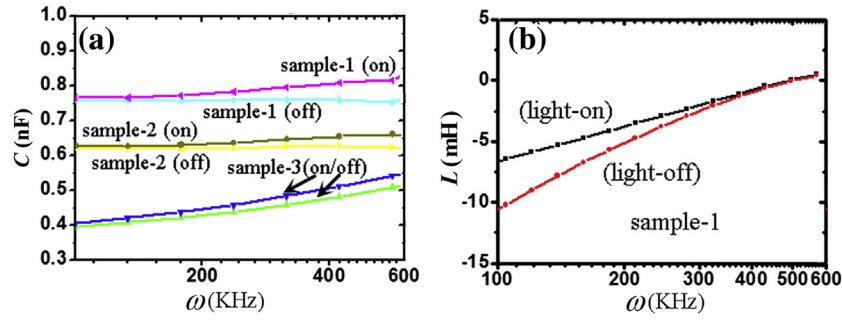


Fig. 4. (a) The C – ω plots obtained from sample 1–3 at illumination-off and -on states, and (b) L – ω plots of sample-1 at illumination-off and -on states.

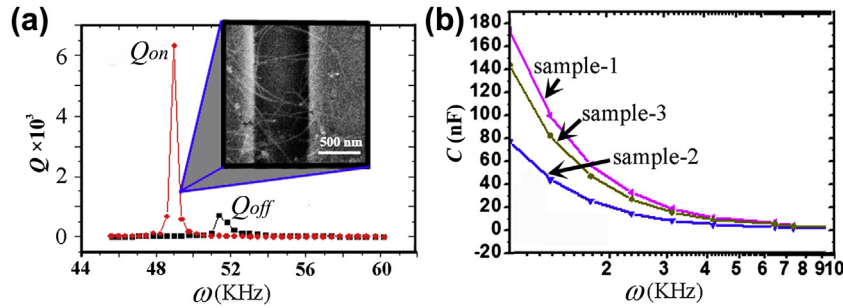


Fig. 5. (a) The Q – ω profile obtained from type-1. Inset: Enhanced SEM image of networked SWCNTs after repeated incidences by laser beam. (b) The C profiles at low ω .

Table 1
Electrical parameters at dark and bright conditions.

	Type 1		Type 2	
	Light-off	Light-on	Light-off	Light-on
ω_o (KHz)	51–55	49–52	152–155	148–151
R (K Ω)	0.104 ± 0.005	0.012 ± 0.001	0.9 ± 0.1	0.76 ± 0.05
Q_{on}/Q_{off}	8		1.2	
R_{off}/R_{on}	8		1.2	

2–3 give $Q_{on}/Q_{off} = 8$ [Fig. 5(a)] and ω_o shift, according to Fig. 4(a) and (b), ranges at 3–5 Hz.

Over 20 samples have been measured and obtained data exhibits a relation of $Q_{on}/Q_{off} \propto R_{off}^{-1}$. For example, devices with low R_{off} give $Q_{on}/Q_{off} > 8$ (Type 1, Table 1) and $1 < Q_{on}/Q_{off} < 2$ emerges as R_{off} is large (Type 2, Table 1); the latter however implies a significant dissipation of I_{photo} . According to the electrical percolation model the equivalent circuit elements of current devices can be expressed as $R_{off} = \Sigma R_n + \Sigma 1/R_n + R_{con}$ where ΣR_n and $\Sigma 1/R_n$

represent nanotube resistors connected in series and parallel respectively and change as tubes are damaged [15]. SEM examination shows that CNTs remain intact after repeated incidences of laser and tube segmentation is absent, indicative of unchanged ΣR_n and $\Sigma 1/R_n$ [inset, Fig. 5(a)]. R_{con} is R arising from tube/lead contacts and controls transmission probability of CNTs [15]. In other words, R_{off} is dominated by R_{con} and equation can be rewritten as $Q_{on}/Q_{off} \propto R_{con}^{-1}$, i.e. the lower the R_{con} the greater the optical gain. Fig. 5(b) verifies R_{con} induced polarization [5] and C at $\omega < 2$ KHz reaches a value as high as 180 nF, corresponding to $X_C = 0.36 \Omega$. Interfacial dissipation is further supported by observations that lighting induces a larger R decrease at reduced T and experiments give 15% at 5 K and 10% at 50 K with respect to Fig. 3(b), indicative of thermalization at tube/lead contacts [Fig. 6(a) and (b)] [17]. Similar results are also observed in 550 nm and 1100 nm excitations and Q_{off}/Q_{on} is measured to be 10.2 and 8.9 at 5 K and, 9.7 and 8.6 at 50 K [Fig. 6(a) and (b)].

Figs 5(b) and 6 clearly indicate that I_{photo} dissipates at tube/lead contacts [Fig. 7(a)] and reduced optical gain accounts for $1 < Q_{on}/$

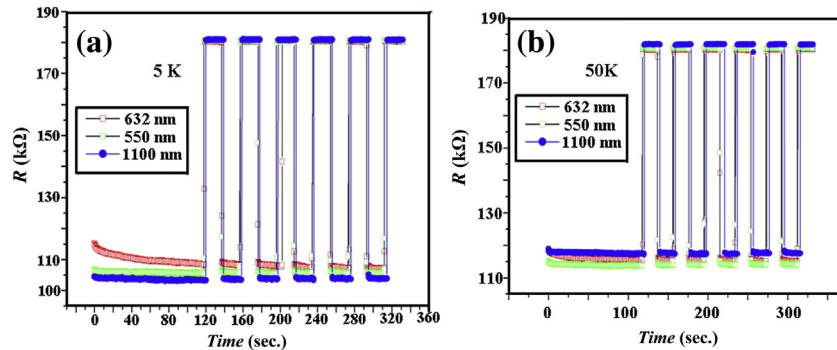


Fig. 6. (a) Time evolved R changes with photo-excitation by various wavelengths at 5 K and (b) 50 K.

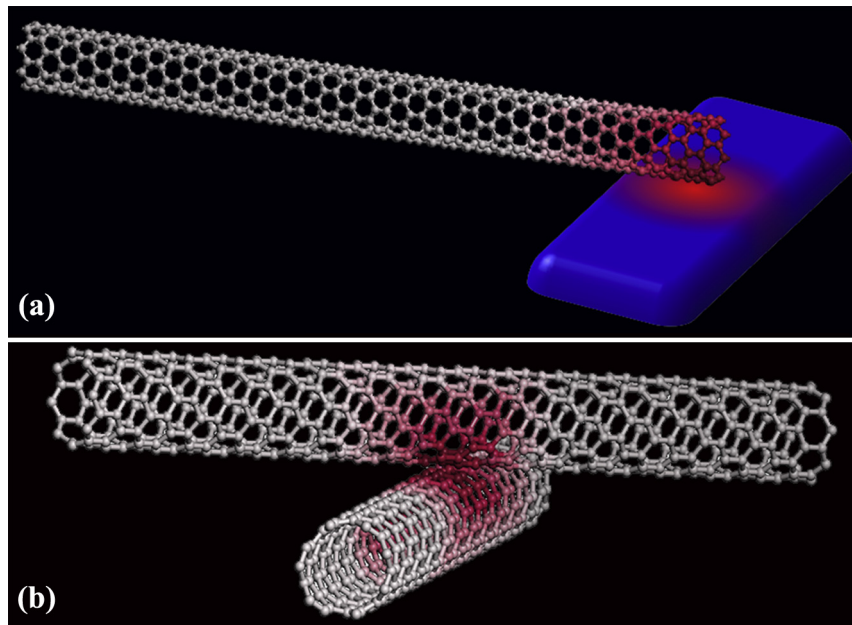


Fig. 7. I_{photo} dissipation at (a) tube-lead contact and (b) intertube junction.

$Q_{off} < 2$ (Type 2, Table 1). For aggregated tubes, excitonic relaxation also takes place at intertube junctions and phenomenon is owing to I_{photo} leakage through surface states, including dipolar lattices and dangling edges [18]. Study reveals that low energy channels are also present in M/S tube junctions [Fig. 7(b)] and E_{SCH} induced hopping is supported by *ab-initio* calculation below. First, S(10,0) and M(9,0) tubes are built in a $1 \times 1 \times 1$ nm cell and tube length is set as 21 nm.

Second, tubes are arranged in a crisscross fashion and three different combinations are computed, including S(10,0)/S(10,0), S(10,0)/M(9,0) and M(9,0)/M(9,0) [Fig. 8(a) and (b)]. Third, structures are geometrically optimized and intertube potential is converged to the van der Waal's regime [18,19]. Fourth, the band diagram is calculated according to density functional theory and charge density is measured with a standard k -point path method [20,21]. Calculation reveals that tubes interact through electronic states near to Fermi level (E_F) [arrows, Fig. 8(c)] and E_{SCH} at S/S and M/M interfaces approaches zero. Transfer however takes place at M/S tube junction and E_{SCH} is calculated to be $3.4 \text{ kcal mol}^{-1}$, corresponding to 1 eV [22]. Fig. 8(d) shows a possible scenario for intertube transfer: first, excitons are created at π^* state (S-tube) and becomes free carriers at $V_{bias} \neq 0$; second, intertube transition of $\pi^*(\text{S-tube}) \rightarrow \pi(\text{M-tube})$ occurs and incommensurate alignment of wave vector at E_F causes carrier scattering at M-tube surfaces. The $\pi^*(\text{S}) \rightarrow \pi(\text{M})$ transition is supported by changeover from negative to positive temperature coefficient of resistivity previously observed in a similar experiment [18]. Yoon et al. have studied the conductance of CNTs at compressive status and data reveal that tunneling proceeds preferentially at deformed lattices. Conductance then drops by two orders of magnitude as load is removed and hopping takes place only at correlated lattices [20]. Crisscrossed tubes, however, cannot electronically correlate and hopping occurs only at the nearest lattices with $E_{SCH} \neq 0$, corresponding to a naphthalene [blue, Fig. 8(b)].

4. Conclusion

SWCNTs exhibit RCL characteristics in ac field and Q is measured to be as high as 1×10^4 . This value can be optically promoted and improvement originates from enhanced C and L upon I_{photo} creation in CNTs. Surface losses account for $1 < Q_{on}/Q_{off} < 2$ I_{photo} and I_{photo} dissipates at tube/lead contacts and S/M tube junctions.

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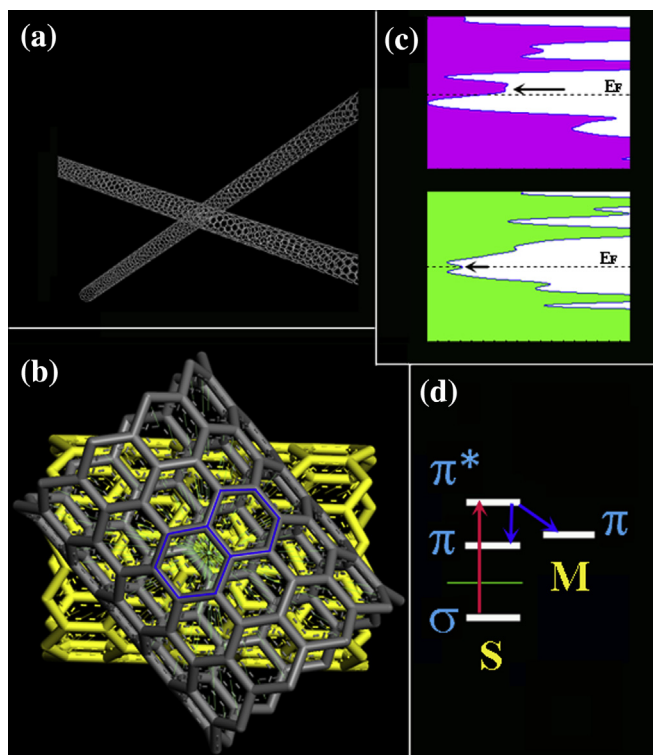


Fig. 8. (a) Simulated structure of crisscross tubes, (b) the nearest lattices with $E_{SCH} \neq 0$ (blue), (c) band diagrams of M- (top) and S-tube (lower) near to the E_F and (d) optical transition from $\pi^*(\text{S})$ to $\pi(\text{M})$.

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